

IN THE UNITED STATE PATENT AND TRADEMARK OFFICE

In re Patent application of

TAKENAKA et al.

Group Art Unit: 1625

5 Appln. No. 10/560,794

Examiner: Covington, Raymond K

Filed: December 15, 2005

Title: Curable Polycyclic Compounds and Process for the  
production Thereof

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Declaration

I Hiromasa YAMAMOTO declare and state that:

1. I completed Department of Science, Graduate School of  
15 Science, Osaka University [Master course] in March of 1987,  
entered Tokuyama Soda K. K. (the name was changed to Tokuyama  
Corporation in 1994) in April of 1987 as a researcher, was  
assigned to Fujisawa Research Laboratory in May of 1987, was  
assigned to Tsukuba Research Laboratories (the name was  
20 changed to Advanced Materials Business Group in 2002) in  
April of 1990, and has worked there until now.

In my assignments in Fujisawa Research Laboratory and  
Tsukuba Research Laboratory, I worked mainly for the  
development of organic materials (in particular, optical  
25 materials and electronic materials) and am versed in the  
technologies thereof.

2. I have reviewed and understand the contents of Dearborn  
DN 48: 13253 (1953) abstract, Ciba DN 57: 75868 (1962)  
30 abstract and the Office Action dated December 26, 2008 in the

subject application.

### 3. Purpose of Experiment

An adamantane compound in which an epoxy-containing  
5 group is introduced at the 1- and 3-positions of adamantane  
ring, can be produced by a method indicated in the present US  
Patent Application No. 10/560,794. The method is a method  
(Claim 19) of first reacting 1,3-adamantanediol with an  
alkali metal hydride to obtain an alcoholate, then reacting  
10 the alcoholate with an allyl halide to obtain 1,3-adamantane  
diallyl ether, lastly oxidizing the allyl group to convert it  
to an epoxy group.

Meanwhile, Dearborn et al. (*Journal of Industrial and Engineering  
Chemistry*(1953),45,2715-21) disclose a method of reacting 1,5-  
15 dihydroxynaphthalene with epichlorohydrin in a presence of  
sodium hydroxide to produce 1,5-bis(2,3-  
epoxypropoxy)naphthalene.

However, the reactivity of the hydroxyl groups directly  
bonded to the 1- and 3-positions of 1,3-adamantanediol are  
20 extremely low as compared with the reactivity of the hydroxyl  
groups of 1,5-dihydroxynaphthalene.

Therefore, when 1,5-dihydroxynaphthalene is replaced by  
1,3-adamantanediol according to the teaching of Dearborn et  
al., no reaction proceeds and it is impossible to produce an  
25 intended 1,3-bis(glycidoxoxy)adamantane compound.

The present experiment is intended to justify the above  
assertion.

### 4. Place where experiment was conducted

30 Tokuyama Corporation, Tsukuba Research Laboratories,

## Experimental Room

40, Wadai, Tsukuba-shi, Ibaraki 300-4247, Japan

## 5. Period of Experiments

5 April 20 to May 12, 2009

## 6. Experiments

I have performed the following experiments.

### Experiment 1

10 In a 100-ml, three-necked flask were placed 10 g (60 mmol) of 1,3-adamantanediol and 22.2 g (240 mmol) of epichlorohydrin. Thereto was dropwise added a 30% aqueous sodium hydroxide solution [1.6 g (0.04 mol) was dissolved in 3.7 g of water]. The mixture was stirred at 80°C for 4 hours.  
15 During the reaction, the amount of 1,3-bis(glycidooxyoxy)adamantane formed was measured by gas chromatography. However, no intended 1,3-bis(glycidooxyoxy)adamantane was detected. After the completion of the reaction, there were recovered 8% of 1-  
20 glycidooxyoxy-3-adamantanol (a mono-substituted product) and 88% of 1,3-adamantanediol (raw material).

In order to enhance the reactivity, there was added, to the above reaction mixture, a 50% aqueous sodium hydroxide solution [1.6 g (0.04 mol) was dissolved in 1.6 g of water],  
25 followed by stirring at 90°C for 6 hours. The reaction was traced by gas chromatography; however, no intended 1,3-bis(glycidooxyoxy)adamantane was detected.

### Experiment 2 The present Invention

30 A method of reacting 1,3-adamantanediol with an alkali metal

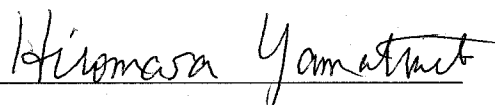
hydride to obtain an alcoholate and then reacting the alcoholate with an allyl halide.

Into a 200-ml, three-necked flask were fed 1.0 g (6.0 mmol) of 1,3-adamantanediol and 20 ml of dimethylformamide. Thereto was added 0.6 g (60 wt. %, 14.3 mmol) of sodium hydride, followed by stirring at 70°C for 2 hours. The reaction mixture was cooled to 5°C. Thereto was dropwise added 2.2 g (18.0 mmol) of allyl bromide. Violent foaming occurred from the timing at which about the half amount was added dropwise. After the completion of the dropwise addition, stirring was conducted at 5°C for 5 hours. 50 ml of water was added and extraction was conducted using 100 ml of methylene chloride. The methylene chloride layer was washed with 20 ml of water four times and the resulting organic layer was subjected to vacuum distillation to distill off the solvent. 1.7 g of a residue was obtained. Analysis of the residue indicated the presence of 94% of 1,3-bis(2-propenyloxy)adamantane. The residue was purified by vacuum distillation to obtain 1.2 g (4.8 mmol) of 1,3-bis(2-propenyloxy)adamantane at a purity of 97%. Of the amount, 1.0 g (4.0 mmol) was fed into a 200-ml, three-necked flask and was dissolved in 50 ml of methylene chloride. Thereto was added 2.50 g (69% product, 10.0 mmol) of m-chloroperbenzoic acid. Stirring was conducted at room temperature overnight. The resulting white precipitate was separated by filtration and the filtrate was washed with 10 ml of a saturated aqueous sodium sulfite solution. The absence of the oxidant in the organic layer was confirmed by a potassium iodide-starch paper. The organic layer was

washed once with a 1 N aqueous sodium hydroxide solution and four times with water. The resulting organic layer was subjected to vacuum distillation to obtain 1.1 g of a residue. Analysis of the residue indicated the presence of 94% of 1,3-  
5 bis(glycidyloxy)adamantane. The residue was purified by vacuum distillation to obtain 1,3-bis(glycidyloxy)adamantane of 97% purity in an amount of 0.9 g (3.2 mmol, yield: 53% (from 1,3-adamantanediol)).

10 I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and believe are believed to be true, and further that these statements are made with the knowledge that willful false statements and the like so made are punishable  
15 by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful statements may jeopardize the validity of the application or any patent issuing thereon.

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Hiromasa YAMAMOTO

Date: May 20, 2009